

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of

Heinrich Becker et al.

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Crouse

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**DECLARATION UNDER 37 C.F.R. '1.132**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Dr. Heinrich Becker, being duly warned, hereby declare that I am an expert in the field of electroluminescent devices, as evidenced by my CV, attached. I am an inventor in the above-captioned application. I performed or supervised the following experiments.

**Experimental Report**

**In the matter of International Patent Application no. PCT/GB03/05523 (Avecia Limited et al)**

**Device Preparation**

An Electroluminescent (EL) device in the form of an Organic Light Emitting Diode (OLED) was prepared in the following manner. The final structure of the device is shown schematically in Figure A attached hereto.

An ITO anode on a glass substrate was used. The ITO anode was of thickness 120nm and roughness 5nm. The anode was cleaned using 4% Deconex (trade name) solution in deionised water followed by ozone treatment.

A hole injection layer (HIL) (thickness = 60nm) comprising an arylamine compound of formula 1 according to the present patent application was deposited onto the ITO anode. In the case of the higher molecular weight arylamine compounds ( $m = 45$ ;  $m = 25$ ), the HIL was deposited by spin coating (from a 4% solution in toluene) followed by drying by heating for 10 min at 110°C. In

the case of the lower molecular weight arylamine compounds ( $m = 1$ ;  $m = 3$ ), the HIL was deposited by evaporation. The rate of deposition by evaporation for all layers in the device was 0.5 nm/s.

For the HIL, the arylamine compounds of formula 1 with  $m = 25$  and  $m = 45$  consisted of poly(2,4 dimethyl triarylamine).

For the HIL, the arylamine compounds of formula 1 where  $m = 1$  and  $m = 3$  were:

$m = 1$	NPB      N,N'-Di(naphthalen-1-yl)-N,N'-diphenylbenzidine (supplied by SynTec, Wolfen, Germany)
$m = 3$	NaphDATA      4,4',4''-Tris(N-(1-naphthyl)-N-phenylamino)triphenylamine (supplied by SynTec, Wolfen, Germany;)

A hole transport layer (HTL) (thickness = 20 nm) was then deposited onto the HIL by evaporation. The HTL comprised spiro-TAD (Tetrakis-2,2',7,7'-diphenylamino-spiro-9,9'-bifluorene).

An electroluminescent layer (EML) (thickness = 30 nm) was deposited by evaporation onto the HTL. The EML comprised either S-DPVBi (2,2',7,7'-Tetrakis-(2,2-diphenyl-vinyl)-spiro-9,9'-bifluorene) ("Sample 1") or a composition of a CBP (4,4'-bis(carbazole-9-yl)-biphenyl) matrix with Ir(piq)<sub>3</sub> (Tris(1-phenyl-isochinolin-C2,N)iridium) as guest material at 20% by weight ("Sample 2").

An electron transport layer (ETL) (thickness = 20 nm) was deposited next by evaporation and comprised Alq<sub>3</sub> (Aluminium-tris-8-hydroxy-chinolat).

Finally, a cathode was deposited by evaporation. The cathode comprised Ba (3 nm) followed by Al (150 nm).

## Results

The measurements (I-V-EI) were made in a conventional way by applying a voltage and measuring the current and brightness. The starting voltage was 0 V and it was increased by steps of 0.2 V until 50 mA/cm<sup>2</sup> were achieved.

### Sample (1):

The results in Table 1 show that Sample (1) when prepared using an arylamine compound of formula 1 ( $m = 45$ ) has a marked improvement in maximum luminescence efficiency (7.2 cd/A) compared to any of the Sample (1) comparisons a-c prepared using arylamine compounds of formula 1 but of lower molecular weight. In addition, the drive voltage is much lower (3.4 V @ 100 cd/m<sup>2</sup>) for the inventive case than for any of the comparative examples. Table 1 also gives data for the colour of the emission of the samples according to the CIE colour system (x and y coordinates), which shows that the colour is hardly changed across the different samples.

### Sample (2):

The results for Sample (2) show the same trends as for Sample (1), i.e. improved maximum luminescence efficiency and lower drive voltage when the device is prepared using an arylamine compound of formula 1 ( $m = 45$ ).

Table 1

Sample	HIL	HTL	EML	ETL	Max efficiency (cd/A)	Drive Voltage (V @100cd/m <sup>2</sup> )	Emission (CIE system)
Sample 1	Formula 1 m = 45	S-TAD	S-DPVBi	Alq	7.2	3.4	x=0.17; y=0.22
Sample 1 (comparison a)	Formula 1 m = 1 (NPB)	S-TAD	S-DPVBi	Alq	3.8	5.8	x=0.17; y=0.25
Sample 1 (comparison b)	Formula 1 m = 3 (NaphDATA)	S-TAD	S-DPVBi	Alq	4.2	5.8	x=0.17; y=0.25
Sample 1 (comparison c)	Formula 1 m = 25	S-TAD	S-DPVBi	Alq	5.3	5.3	x=0.17; y=0.24
Sample 2	Formula 1 m = 45	S-TAD	CBP : Ir(piq) <sub>3</sub> (20 %)	Alq	5.5	5.2	x=0.31; y=0.69
Sample 2 (comparison a)	Formula 1 m = 1 (NPB)	S-TAD	CBP: Ir(piq) <sub>3</sub> (20 %)	Alq	4.4	6.5	x=0.31; y=0.69
Sample 2 (comparison b)	Formula 1 m = 25	S-TAD	CBP: Ir(piq) <sub>3</sub> (20 %)	Alq	5.1	5.9	x=0.31; y=0.69

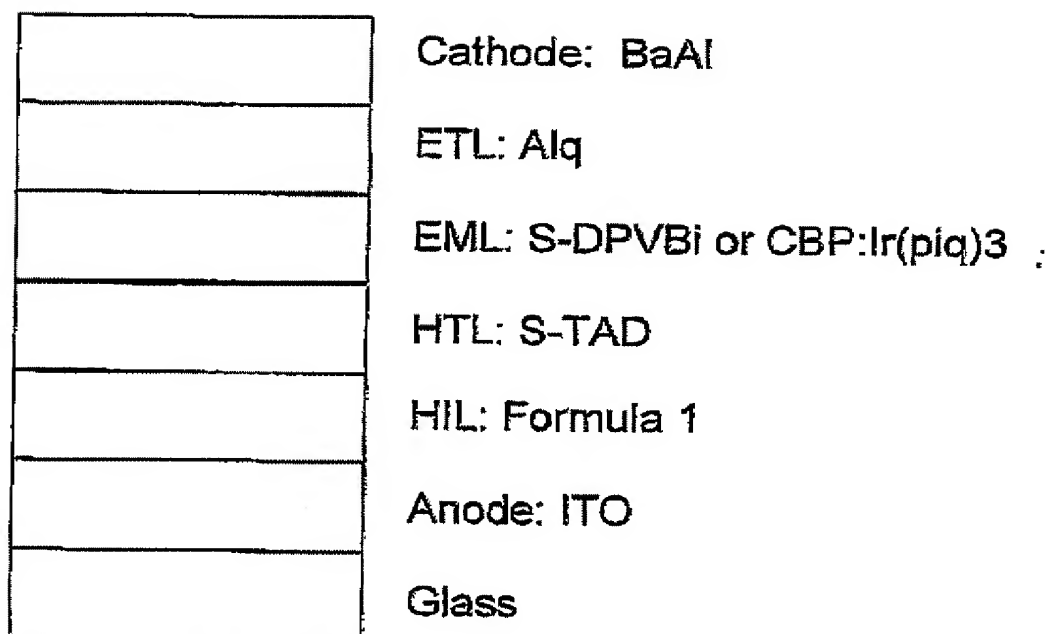


Figure A

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Sept. 25<sup>th</sup> 2009  
Date

Dr. Hein Rens

# Dr. Heinrich Becker

## Curriculum Vitae

Name: Dr. Heinrich Becker  
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65719 Hofheim  
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### Work Experience:

2005–present Research, scaling and production of Light Emitting Polymers at Merck KGaA.  
1999–2005 Manager Research & Development at Covion Organic Semiconductors GmbH.  
1998–1999 Laboratory manager at Covion Organic Semiconductors GmbH.  
April 1998 Inaugural member of the management team of Covion Organic Semiconductors GmbH.  
1995–1998 Laboratory manager at the corporate research of Hoechst AG (Light Emitting Organics project).

### Academic Qualification:

1993–1995 Postdoctoral Stay with Prof. Dr. K. Barry Sharpless, The Scripps Research Institute, La Jolla, USA.  
1990–1993 Doctor of Philosophy (Dr. phil. nat.) Organic Chemistry, Johann Wolfgang Goethe University Frankfurt, Dept. of Organic Chemistry, Prof. Dr. Gerhard Quinkert, Thesis title: "Stereoselective reactions on macrolides".  
1990–1992 Inaugural Member of the Frankfurt-Graduiertenkolleg "Chemical and biological synthesis of pharmaceuticals".  
1989–1990 Graduate as "Diplom-Chemiker", Johann Wolfgang Goethe-University Frankfurt, Dept. of organic chemistry, Prof. Dr. Gerhard Quinkert, Thesis title: "The enantioselective introduction of the stereogenic center C(17) at (+)-Aspicilin".  
1985–1989 Undergraduate studies at the Johann Wolfgang Goethe-Frankfurt.